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## **Report Title**

Frequency Conversion of Short Optical Pulses in Negatively Spatially Dispersive Metamaterials

### **ABSTRACT**

We show that particular spatial distributions of nanoscopic plasmonic building blocks in metamaterials may enable extraordinary nonlinear-optical frequency-shifted reflectivity and pulse shaping.

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# Frequency Conversion of Short Optical Pulses in Negatively Spatially Dispersive Metamaterials

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**Abstract:** We show that particular spatial distributions of nanoscopic plasmonic building blocks in metamaterials may enable extraordinary nonlinear-optical frequency-shifted reflectivity and pulse shaping.

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Metamaterials (MM) are artificially designed and engineered materials, which can have properties unattainable in nature. Usually, MM rely on advances in nanotechnology to build tiny metallic nanostructures smaller than the wavelength of light. These nanostructures modify the electromagnetic (EM) properties of MM, sometimes creating seemingly impossible optical effects. Negative-index MM (NIMs) are most intriguing EM materials that support backward EM waves (BEMW). Phase velocity and energy flux (group velocity) become *contra-directed* in NIMs. The appearance of BEMW is commonly associated with simultaneously negative electric permittivity ( $\epsilon < 0$ ) and magnetic permeability ( $\mu < 0$ ) at corresponding frequencies and, consequently, with negative refractive index  $n = -\sqrt{\mu\epsilon}$ . Counter-intuitive backwardness of EMW is in strict contrast with the electrodynamics of ordinary, positive-index materials. The possibility of huge enhancement of optical parametric amplification has been predicted through three-wave mixing in NIMs if one of the coupled electromagnetic waves falls in the negative-index frequency domain [1,2]. Unparalleled properties of second harmonic generation in NIMs were predicted in [2,3]. It was shown that frequency-mixing of ordinary and BEMV enables frequency shifted amplified reflection of electromagnetic waves [4].

Current mainstream in fabricating bulk NIM slabs relies on engineering of LC nanocircuits - plasmonic mesoatoms with negative electromagnetic response. Extraordinary coherent nonlinear optical frequency-converting propagation processes predicted in NIMs have been realized (simulated) so far only in the microwave transmission lines [5]. This paper proposes a different paradigm which employs spatial dispersion [6] to realize outlined processes. We show that metamaterial slabs which may support coexistence of ordinary electromagnetic waves with co-directed phase velocity and energy flux ( $\partial\omega/\partial k > 0$ ) and extraordinary optical modes with contra-directed phase and group velocities ( $\partial\omega/\partial k < 0$ ) enable extraordinary nonlinear optical processes similar to that predicted in NIMs. Particularly, the focus is on the extraordinary properties of nonlinear optical frequency-mixing of contra-propagating ordinary short optical pulses and pulses with negative group velocity which have been studied through numerical simulations.

One of the possible realization of the outlined approach is a plasmonic metaslab made of carbon nanotubes. It can be viewed as a plasmonic wave guide formed by a metal plate (bottom) and by air (top) tampered by standing carbon nanotubes [7]. Such a structure supports different EM eigenmodes in the THz and mid IR. Some of them exhibit spatial dispersion which changes from positive to negative in different wavelength intervals whereas others predominantly maintain negative dispersion. Frequencies and gaps between the modes can be tailored by changing lengths and spacing between the nanotubes so that phase velocities for the modes matching the photon energy conservation law would become equal as their group velocities remain contra-directed. Basically, different materials and geometries can be utilized to control spatial dispersion and losses in such MM with variable spatial dispersion [8].

Unparalleled properties of nonlinear-optical coupling (collision) of short contra-propagating pulses in such MMs are illustrated in Fig. 1 (a) – (d) for the example of BW second harmonic (SH) generation in the loss-free MM. Here  $\tau$  is the time interval measured in units of the input pulse duration [Figs. 1 (a), (b)]. Position of the fundamental and SH pulses inside the metaslab is measured in units of the input pulse length  $l$  [Figs. 1 (c), (d)]. Second harmonic is generated only inside the fundamental pulse. It travels in the opposite direction and exits from the fundamental pulse. This results in different durations of the fundamental and SH pulses exiting from the opposite sides of the

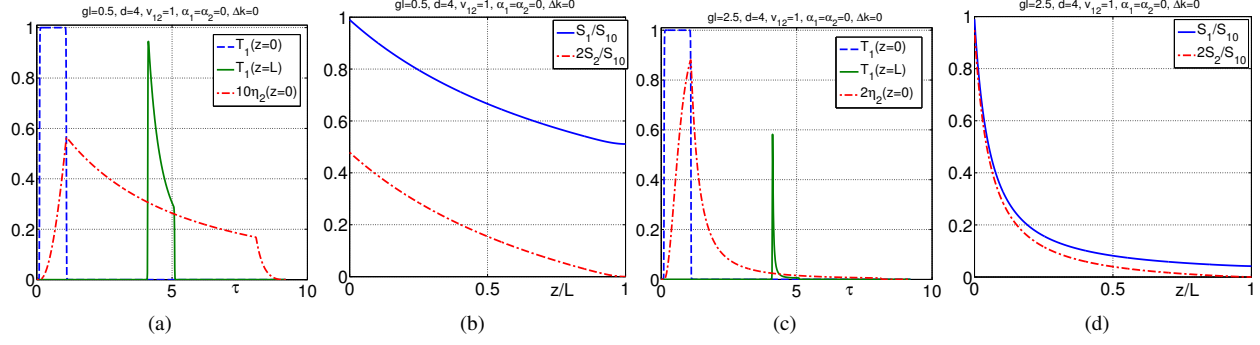


Fig. 1. Phase-matched backward-wave SH generation in the loss-free MM. (a) and (c): Input rectangular  $T_1(z=0)$  and output  $T_1(z=L)$  pulse shapes for the fundamental radiation;  $\eta_2(z=0)$  are pulse shapes for the emitted back second harmonic. (b) and (d): Change of the pulse energy at the corresponding frequencies across the slab. Here,  $d = L/l$ ,  $2S_2/S_{10}$  is photon conversion efficiency.  $\alpha_{1,2}$  and  $g$  are absorption and nonlinear coupling parameters.

metaslab. The metaslab operates as controlled frequency doubling microscopic nonlinear-optical metamirror. Shapes of the output transmitted and generated SH pulses depend on the energy conversion rate inside the fundamental pulse and, hence, can be controlled by changing intensity and ratio of the input pulse length to the metamaterial thickness (parameter  $d$ ). It is demonstrated in Figs. 1 (a), (b). Figures 1 (c), (d) display overall pulse energy (photon) conversion efficiency across the metaslab. Unusual behavior is due to the opposite propagation direction of the pulses.

In the conclusion, we show that a deliberately engineered highly dispersive metamaterial slab enables switching of optical pulses to contra-propagated frequency shifted pulses with tailored pulse shape. Enhanced efficiency of switching is ensured by the extraordinary phase matching of contra-propagating light pulses. Extraordinary properties of coherent nonlinear-optical switching in short-pulse regime in such metamaterials are demonstrated through numerical simulations. It is shown that greatly enhanced nonlinear optical coupling and frequency conversion become possible due to extraordinary opposite directions of phase and group velocities for one of the pulses, whereas other coupled pulses remain ordinary. Then the metaslab operates as a *frequency-converting nonlinear-optical microscopic metamirror and switch*. Other prospective applications in the photonic device technologies will be discussed.

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